Atmospheric CO2 modeling at the regional scale: Application to the CarboEurope Regional Experiment

Sarrat, C.; Noilhan, J.; Lacarrere, P.; Donier, S.; Lac, C.; Calvet, J.C.; Dolman, A.J.; Gerbig, C.; Neininger, B.; Ciais, P.; Paris, J.D.; Boumard, F.; Ramonet, M.; Butet, A.

published in
Journal of Geophysical Research
2007

DOI (link to publisher)
10.1029/2006JD008107

document version
Publisher's PDF, also known as Version of record

Atmospheric CO_2 modeling at the regional scale: Application to the CarboEurope Regional Experiment

C. Sarrat,[1] J. Noilhan,[1] P. Lacarrère,[1] S. Donier,[1] C. Lac,[1] J. C. Calvet,[1] A. J. Dolman,[2] C. Gerbig,[3] B. Neininger,[4] P. Ciais,[5] J. D. Paris,[5] F. Boumard,[5] M. Ramonet,[5] and A. Butet[6]

Received 5 October 2006; revised 27 November 2006; accepted 12 February 2007; published 16 June 2007.

[1] The CarboEurope Regional Experiment Strategy (CERES) experiment took place in May and June 2005 in France and offers a comprehensive database on atmospheric CO_2 and boundary layer processes at the regional scale. One "golden" day of CERES is interpreted with the mesoscale atmospheric model Meso-NH coupled on-line with the Interactions between Soil, Biosphere and Atmosphere, CO_2-reactive (ISBA-A-gs) surface scheme, allowing a full interaction of CO_2 between the surface and the atmosphere. The rapid diurnal cycle of carbon coupled with water and energy fluxes is parameterized including, e.g., plant assimilation, respiration, anthropogenic emissions, and sea fluxes. During the analyzed day, frequent vertical profiles and aircraft transects revealed high spatial and temporal variabilities of CO_2 concentrations within the boundary layer at the regional scale: a 10-ppm gradient of CO_2-mixing ratio is observed during the day by the aircraft measurements. The Meso-NH model proved able to simulate very well the CO_2 concentration variability as well as the spatial and temporal evolution of the surface fluxes and the boundary layer in the domain. The model is used to explain the CO_2 variability as a result of two complementary processes: (1) the regional heterogeneity of CO_2 surface fluxes related to the land cover (e.g., winter crops versus a pine forest) and (2) the variability of mesoscale circulation across the boundary layer: development of the sea breeze in the western part of the domain and dominating wind flow in the eastern part of the domain.

Citation: Sarrat, C., et al. (2007), Atmospheric CO_2 modeling at the regional scale: Application to the CarboEurope Regional Experiment, *J. Geophys. Res.*, *112*, D12105, doi:10.1029/2006JD008107.

1. Introduction

[2] The atmospheric carbon dioxide (CO_2) measurements provide key information to improve our understanding on the terrestrial carbon budget. An important issue of the carbon cycle is to improve the retrieval of sources and sinks by inverse modeling [*Rayner et al.*, 1999; *Bousquet et al.*, 1996; *Gurney et al.*, 2002].

[3] At the regional scale, vegetation carbon uptake and ecosystem respiration, in combination with diurnal variations in vertical mixing, result in a pronounced diurnal cycle of CO_2 in the atmospheric boundary layer (ABL). Even at the continental scale, *Denning et al.* [1996] or *Chevillard et al.* [2002] showed the importance of the diurnal cycle of biospheric exchanges and of regional transport to take into account the short-term variations in the CO_2 concentrations.

[4] Preliminary studies with high resolution mesoscale models are reported by, e.g., *Nicholls et al.* [2004] and *Denning et al.* [2003], who showed that the mesoscale Regional Atmospheric Modeling System (RAMS) model, coupled with the biospheric SiB-2 model, was able to retrieve the CO_2 flux and concentration measurements made on a tall tower at various heights. The study of *Geels et al.* [2004] describes a mesoscale CO_2 simulation to investigate the CO_2 synoptic variability with a relatively large spatio-temporal resolution (daily, $1° \times 1°$).

[5] In order to improve our knowledge and the models at the regional scale, a regional experiment has been executed in 2005 in the southwest of France: CarboEurope Regional Experiment Strategy (CERES), within the frame of the European project CarboEurope-IP. Campaigns on regional CO_2 have been held before {Regional Assessment and Monitoring of the Carbon Balance within Europe (RECAB), COCA, *Schmitgen et al.* [2004], COBRA, *Gerbig et al.* [2003]}, but

[1]Centre National de Recherches Météorologiques/Groupe d'études de l'Atmosphère Meteórlogique, Météo France, Toulouse, France.
[2]Vrije University, Amsterdam, Netherlands.
[3]Max Planck Institute for Biochemistry, Jena, Germany.
[4]MetAir, Menzingen, Switzerland.
[5]Le Laboratoire des Sciences du Climat et l'Environnement, Gif sur Yvette, France.
[6]Service des Avions Français Instrumentés pour la Recherche en Environnement Météo France, Toulouse, France.

0148-0227/07/2006JD008107

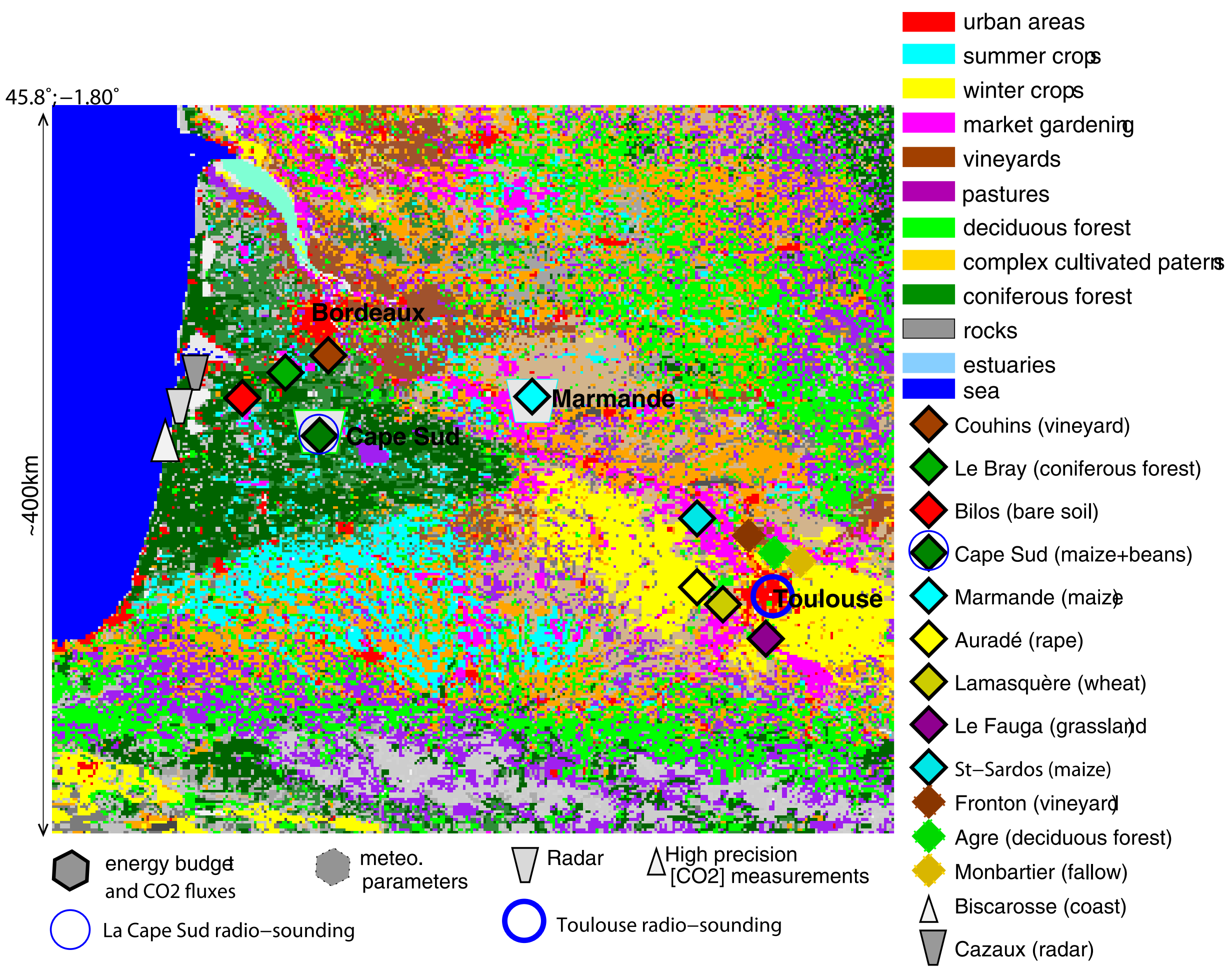

Figure 1. Map of surface vegetation cover over the CERES domain with experimental network (black squares are flux stations).

CERES has a longer temporal coverage (6 weeks) and a much denser sampling of both atmospheric concentrations and of ecosystem fluxes [*Dolman et al.*, 2006].

[6] This paper summarizes a high-resolution modeling study of an episode of the CERES campaign. This is a so-called "golden" day that presents unexpected regional distribution of CO_2 concentrations with strong heterogeneities in the ABL.

[7] In order to separate the contribution of the different physical processes (fluxes versus transport processes) involved in controlling the spatiotemporal CO_2 evolution, the distribution of atmospheric CO_2 this day is simulated with a high resolution mesoscale meteorological and transport model.

[8] After a description of the experimental and modeling configurations, the modeling results, in comparison with the observations, are discussed and used to explain the roles of mesoscale circulations and surface exchanges in the regional pattern of CO_2, in particular the contrasts between forest and crop covered landscapes.

2. The CarboEurope Regional Experiment Strategy: CERES

[9] The CarboEurope Regional Experiment Strategy (CERES) campaign too ce in the southwest of France in May and June 2005 [*Dolman et al.*, 2006], over an area of 300 × 300 km, including the Landes forest, winter and summer croplands areas, the cities of Bordeaux in the northwest part of the domain, and Toulouse in the southeast part. The studied domain is contained in Figure 1 representing the ECOCLIMAP land cover by *Champeaux et al.* [2005].

[10] The aim of CERES was to determine the regional balance of carbon over the southwest of France and to provide a rich database on CO_2 concentrations, surface fluxes, and atmospheric conditions to quantify the carbon budget.

[11] This region has been chosen because of its geographical location: the Atlantic Ocean at its western boundary (expectedly offering incident "well-mixed" air masses at the domain's boundary), a large and homogeneous area of pine forest (Les Landes), and agricultural areas at the eastern part of the domain. The region is only slightly affected by anthropogenic emissions of CO_2 with the exception of the two large cities of Bordeaux and Toulouse. Moreover, this region is very well known in terms of meteorology and hydrology because of past experimental efforts in the area (e.g., the Hapex-Mobilhy experiment [*André et al.*, 1986; *Noilhan et al.*, 1991; *Habets et al.*, 1999].

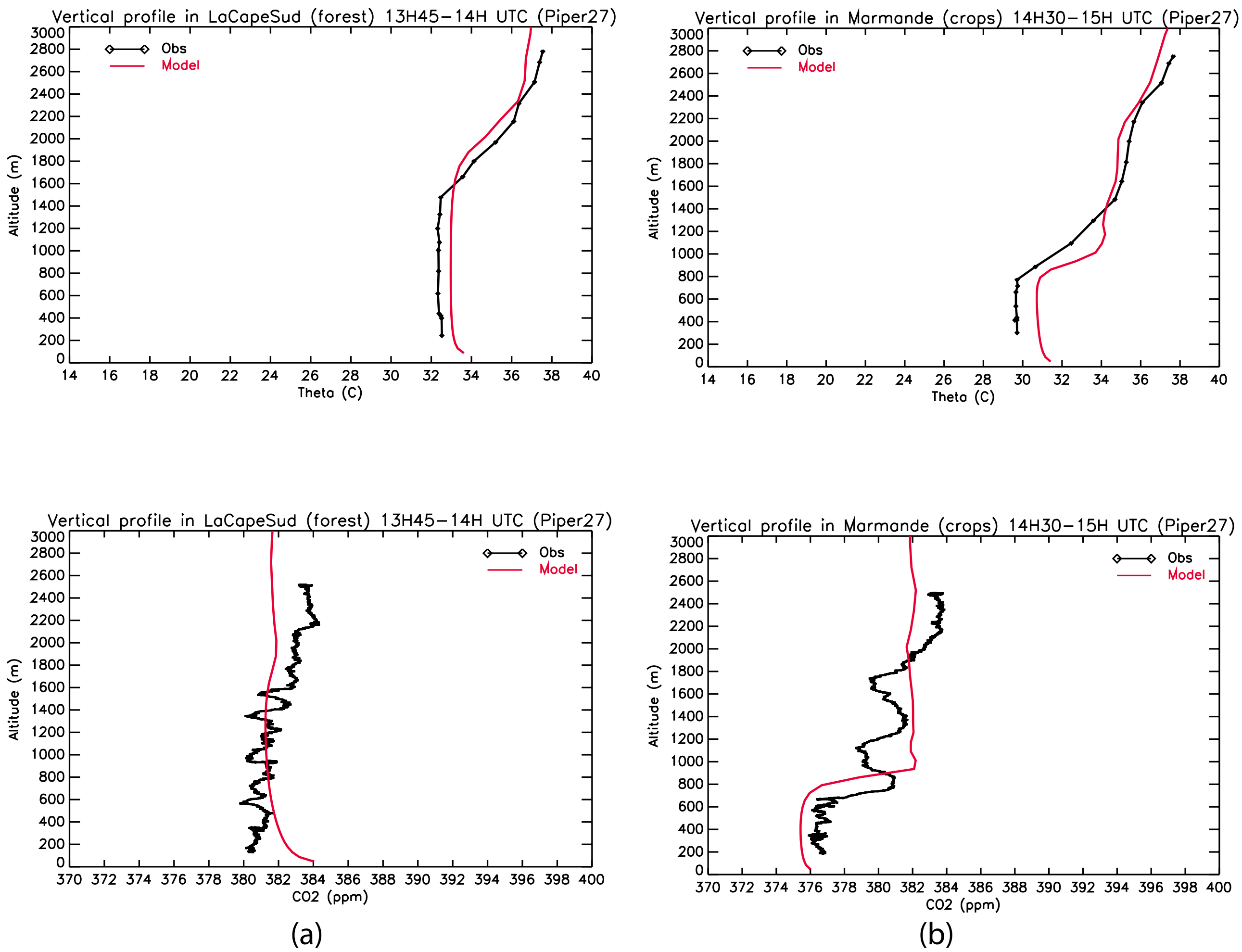

Figure 2. Comparisons between observed (Piper Aztec aircraft) and simulated vertical profiles of CO_2 concentrations (top) and potential temperature (bottom) (a) over La Cape Sud and (b) over Marmande.

[12] A total of 10 eddy covariance flux stations gave continuous measurements of energy, water, and CO_2 fluxes over different types of vegetation (maize, winter crops such as wheat and rapeseed, forest, grassland, vineyard, beans, and clear-cut). A central site was based in La Cape Sud in Les Landes forest, with energy and CO_2 flux measurements, intensive radio-sounding launches during Intensive Observation Period (IOP), and an ultrahigh frequency (UHF) radar for wind measurement (Figure 1). A secondary site was based in Marmande, in the center of the domain dominated by cropland, which included a tower equipped to measure CO_2 concentrations at five levels up to 20 m. A 50-m tower was based in the Atlantic Ocean shoreline in Biscarosse to measure continuously the CO_2 concentrations.

[13] Four research aircraft were operated during IOP. These instrumented aircraft sampled the horizontal and temporal heterogeneities of the CO_2 distribution and fluxes at the regional scale in the morning and afternoon. The aircraft also sampled the heterogeneity in and above the ABL, executing vertical profiles above the different sites (La Cape Sud, Marmande, and shoreline). The in situ CO_2 measurements used in this study come from the Piper-Aztec and the Eco-Dimona air In the Piper-Aztec, the CO_2 is measured by infrared absorption (CONDOR system), and aboard the Eco-Dimona, the measurements were made using a closed-path licor, an open path licor, and flask analysis. CO_2 in situ data have been adjusted to match the measurements of simultaneously collected flask samples by the two aircraft and thus are not regarded as independent measurements.

[14] This study focuses on 26 and 27 May (hereafter referred as may-26 and may-27) because this period is intensively documented. Anticyclonic weather conditions prevailed (clear sky, high temperatures, and light winds), which favored the development of breeze circulations.

3. The Modeling Configuration

[15] The simulation is performed with the Meso-NH atmospheric model [*Lafore et al.*, 1998] coupled on-line with the ISBA surface scheme [*Noilhan and Planton*, 1989] in its carbon interactive mode [*Calvet et al.*, 1998]. Meso-NH is a nonhydrostatic mesoscale atmospheric model allowing two-way nesting simulations. The chosen configuration for Meso-NH is composed of two nested domains at 10-km resolution for the larger one (width, 900 × 900 km)

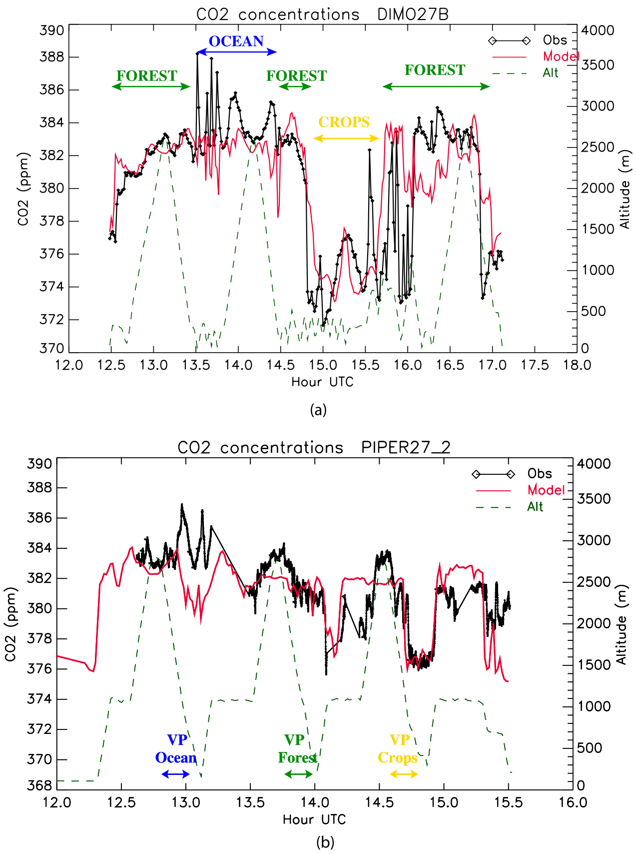

Figure 3. Temporal evolution of CO_2 concentrations measured and simulated (a) during the Eco-Dimona flight and (b) during the Piper Aztec flight (VP, vertical profile). Aircraft trajectories in Figure 4d. The CO_2 concentration comparisons are done at the exact time and location.

and 2-km resolution for the small one (width, 320 × 320 km included in Figure 1). The vertical grid, which is composed of 57 levels, has a higher resolution close to the surface, with 20 levels between 0 and 1000 m, in order to obtain a detailed description of the ABL. The simulation of the CERES IOP2 starts on may-26 at 1800 Coordinated Universal Time (UTC) and continues until may-28 at 0000 UTC. The model is initialized and forced at the lateral boundaries every 6 hours by the European Centre for Medium-Range Weather Forecasting (ECMWF) large-scale analyses for atmospheric and surface variables (the initialized soil moisture was found close to the observations). The CO_2 concentrations are initialized with a homogeneous vertical profile all over the domain at 1800 UTC on may-26, issued from aircraft observations. At the lateral boundaries, a zero-gradient condition is applied for CO_2. The surface energy and CO_2 fluxes are computed on-line by the surface model, ISBA-A-gs, including vegetation assimilation and respiration [*Calvet et al.*, 1998]. A tile approach is used to take full advantage of the 1-km resolution vegetation database. The land cover is from the ECOCLIMAP database [*Champeaux et al.*, 2005; *Masson et al.*, 2003]. It contains 62 types of cover and discriminates winter crops from summer crops (see Figure 1). The oceanic CO_2 fluxes are estimated according to *Takahashi* [1997] using simulated winds for air-sea gas exchanges. The anthropogenic sources come from the 10-km resolution inventory of the Stuttgart University [*Dolman et al.*, 2006].

4. Results

[16] The studied day (may-27) is warm; air temperature in Marmande reaches 31.82°C. In the morning, the anticyclonic conditions drive a weak southeasterly wind at low levels. During the afternoon, the Piper Aztec flew over the Landes forest to Marmande and landed in Toulouse. During the flight, two vertical profiles were made: the first one over the forest (La Cape Sud) at 1400 UTC and the second one over the maize cropland (Marmande) half an hour later. These observed vertical profiles are illustrated in Figure 2, together with the simulated ones (all the modeling results are provided by the 2-km resolution model). The vertical profile of potential temperature is well reproduced by the model over both sites: the boundary layer height is 1600 m in La Cape Sud and only 800 m in Marmande, both in data and model. The differences of ABL height are due to large differences in the surface sensible heat flux, in agreement with observations (not shown). The sensible heat flux is high over the forest, whereas in the eastern agricultural area, the sensible heat flux is lower. In the agricultural area, the latent heat flux is high where the winter crops are fully developed and low in the summer crop areas (mainly maize) dominated by bare ground at this period of the year. For this land cover, the sensible heat is reduced because of high heat storage in the ground.

[17] The temperature vertical profile discrepancies between the model and the observations do not exceed 2°C in and above the ABL.

[18] The comparisons between observed and simulated CO_2 mixing ratio vertical profiles over La Cape Sud can be considered in good agreement in the well-mixed ABL even if there is a 4-ppm discrepancy near the surface. The observations indicate that there is a net CO_2 sink near the surface (inferred from the gradient in CO_2, Figure 2a), but the model results show an opposite gradient. In the model, because of an excessive ecosystem respiration, the net CO_2 in the afternoon tends to be slightly overestimated. This is a component of the surface scheme which needs to be improved, particularly the dependence between the ecosystem respiration and the surface temperature. In Marmande, the comparisons are in good agreement in all the ABL. These observed profiles at the two sites are well mixed in the ABL with a sharp gradient at the capping inversion, stronger in the Marmande case.

[19] What is remarkable in these two punctual observations is that the mean CO_2 mixing ratios in the boundary layer are different at the two sites: 377 ppm in Marmande and 381 ppm in La Cape Sud, that is to say a significant difference between these two selected sites separated by 70 km only.

[20] These differences in CO_2 concentrations are even emphasized between the forest area and the cropland as measured during the Eco-Dimona flight (Figure 3a) over a distance of 750 km: a 10-ppm horizontal gradient of CO_2 mixing ration exists between these two zones (the aircraft trajectory is plotted in blue in Figure 4d). The observed and modeled CO_2 concentrations show a remarkably good

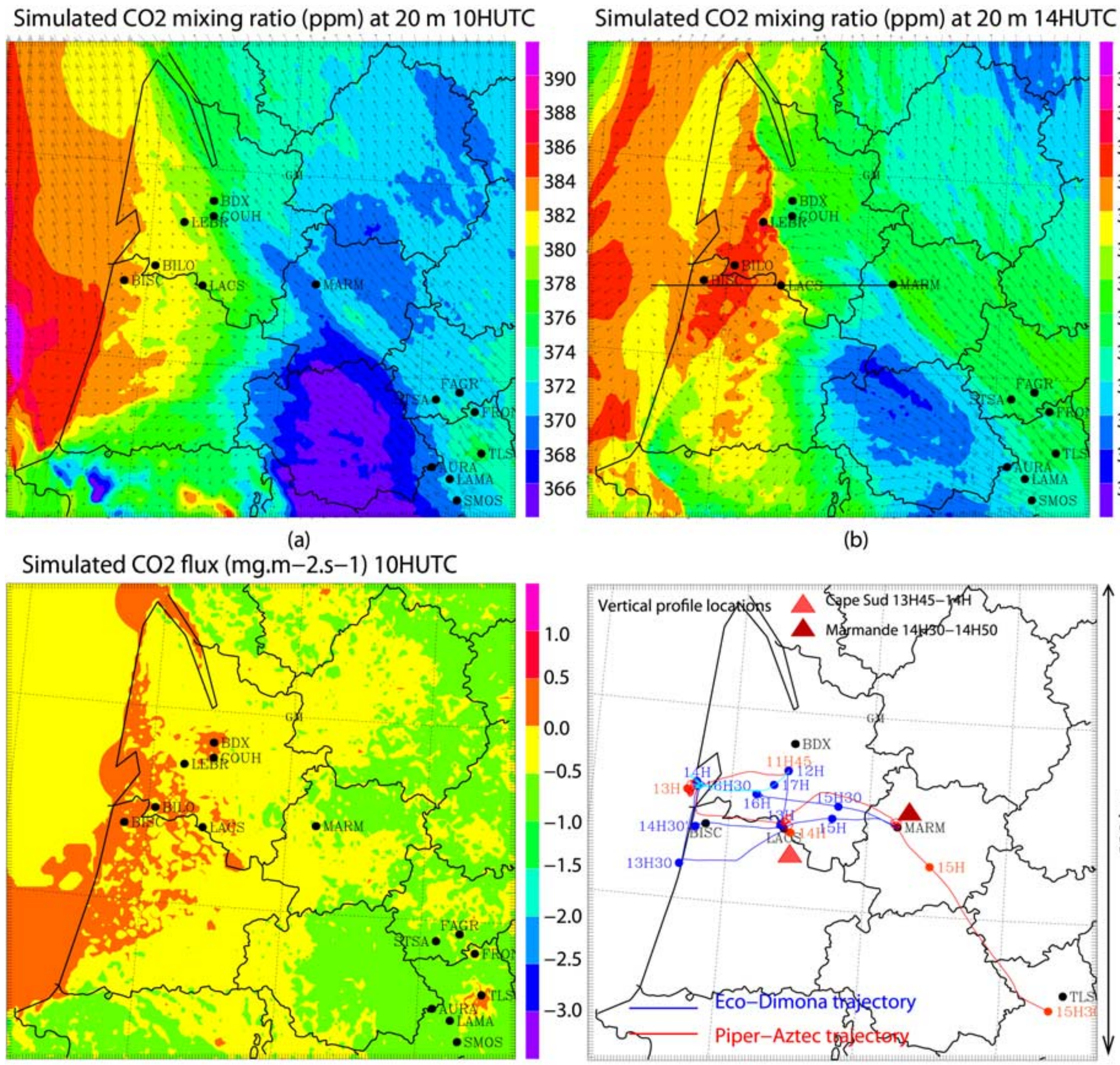

Figure 4. Simulated two-dimensional fields of: (a) CO_2 concentrations with wind arrows at the first level of the model (i.e., 20 m) at 1000 UTC and (b) 1400 UTC; the black line represents the vertical cross section shown in Figure 5; (c) CO_2 surface fluxes (mg m^{-2} s^{-1}) at 1000 UTC and (d) aircraft trajectories: in blue, the Eco-Dimona and in red, the Piper Aztec.

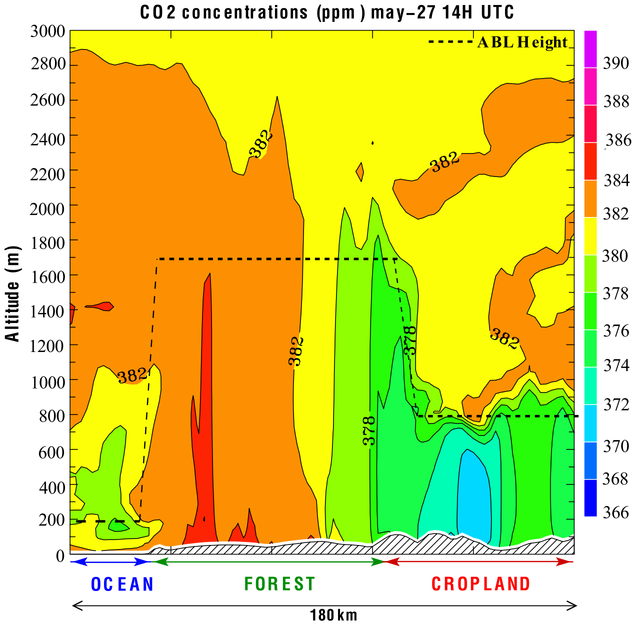

Figure 5. Vertical cross section of simulated CO_2 concentrations (ppm) on may-27 at 1400 UTC. The dotted line represents the approximative height of the atmospheric boundary layer.

agreement in the ABL, and this is during the 5 hours of the flight path. The model is able to reproduce the CO_2 spatial and temporal heterogeneity at the regional scale. In fact, when the aircraft flies over the forested area or near the ocean shoreline, i.e., between 1230 and 1500 UTC, the air sampled is rich in CO_2 (up to 386–388 ppm), and when it flies over the agricultural area, i.e., between 1500 and 1530 UTC over Marmande, the air is more depleted, with mixing ratios of down to 372 ppm, measured and also simulated.

[21] The CO_2 gradient between the forest and the agricultural area is also sampled by the Piper Aztec aircraft (Figure 3b) on a larger horizontal distance. The CO_2 concentrations slowly decrease from the western part of the domain to the eastern part on a distance of 200 km, both in the model and in the observations. Once again, the model reproduces the slight variations of CO_2 at the scale of a few hundred kilometers.

[22] This CO_2 difference between these two ecosystems are due to several processes occurring during this day. The mesoscale modeling is an efficient tool to clearly show that, during the morning, the dominant wind direction in the CERES region is from southeast (see Figure 4a). The southeast of the domain is mainly cultivated with winter crops (yellow on the map, Figure 1). This vegetation type assimilates a larger amount of CO_2 compared to other vegetation types. The uptake of CO_2 in the morning and in the afternoon is higher in this area than anywhere else as seen in Figure 4c. The winter crop uptake creates a CO_2 depletion (well shown in Figure 4b) in the shallow morning ABL that is advected to the northwest toward Marmande. On the other hand, in the beginning of the afternoon, a sea breeze is developing and penetrates a few tens of kilometers inland from the southwest. Over the forest, high CO_2 concentration is assoc with the wind convergence between the westerly sea breeze and a southeast wind in the eastern part (Figure 4b).

[23] The CO_2 gradient between the forest and the agricultural area, together with the ABL height differences between these two zones, are clearly simulated as shown in the vertical cross section of CO_2 concentrations displayed in Figure 5. This vertical cross section of CO_2 is along the black line represented in Figure 4b, from the ocean to Marmande, running through La Cape Sud. Although the ABL in La Cape Sud reaches up to 1600 m, the vertical mixing of CO_2 into the ABL is not sufficient to decrease the concentrations that remain higher than in Marmande, where the ABL reaches only 800 m between 1400 and 1500 UTC.

[24] The wind convergence and the difference of surface fluxes between the forest and winter or summer crops are the two factors that generate a 10-ppm horizontal gradient between the forest and the cropland. This hypothesis is confirmed by the budget calculations made with the model. During the morning and until 1100 UTC, the entrainment at the top of the boundary layer is negligible, while the surface fluxes of CO_2 are identical at the two sites (forest and cropland). Over the forest, the advective tendencies are very weak for CO_2 concentrations and slightly positive (advection of air rich in CO_2), while over the summer cropland, the negative advective tendencies pilot the CO_2 concentration evolution (loss of CO_2 by advection of air depleted in CO_2 and coming for the area covered by winter crops in the eastern part of the domain). In the morning, the advection process represents, in Marmande, a loss of up to 4 ppm of CO_2 per hour.

[25] Figure 6 gives a schematic representation of the main physical processes along a west-east vertical cross section.

5. Conclusion

[26] The CERES experiment conducted in the southwest of France in May and June 2005 offers an adequate and comprehensive database for mesoscale atmospheric modeling of carbon dioxide. It reveals interesting regional conditions in which the geographic distribution of CO_2 is mainly driven by the surface exchanges and atmospheric boundary layer processes and also regional circulations. During one "golden" day of CERES, the observations of two research aircraft exhibit a large gradient of CO_2 mixing ratio with a difference of mean 10 ppm between the forest and the agricultural area over a distance of 70 km in the boundary layer for several hours.

[27] The meteorological model Meso-NH, coupled with the ISBA-A-gs surface scheme and driven by a very precise land cover map, reproduces the gradient of CO_2 very accurately, as well as the ABL vertical profile of potential temperature.

[28] The model proved to be an efficient tool for understanding the processes involved in the observed CO_2 heterogeneity. In fact, the model shows a special pattern of regional circulation with (1) at the eastern part of the domain, a southeast flow that advects low CO_2 concentration in air exposed to winter crop assimilation and (2) at the western part of the domain, higher CO_2 concentrations that accumulate over the forest because of a lower assimilation (the forest becomes a source during the afternoon) and the sea breeze advecting higher CO_2 concentrations.

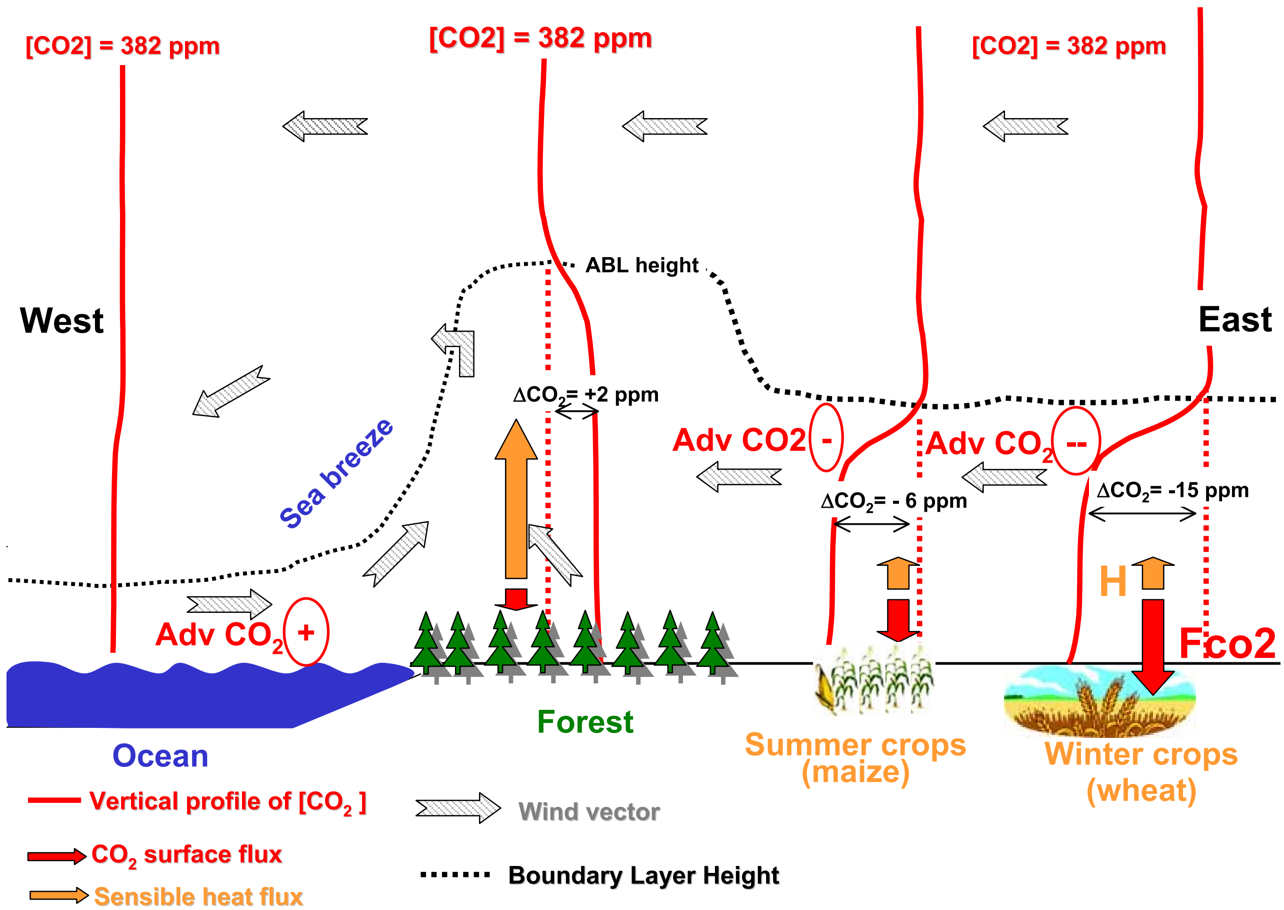

Figure 6. Schematic description of the main physical processes along a vertical west-east cross section on may-27 around 1400 UTC: the high ABL above the pine forest is due to a high sensible heat flux. The CO_2 concentration slightly increases in the ABL due to advection of CO_2 by the sea breeze and because of a small CO_2 surface flux. The ABL height decreases over the eastern crops where the sensible heat is weak. The CO_2 concentrations in the ABL decreases remarkably over the winter crops area characterized by a high assimilation rate. Over the summer crops, despite a relatively small assimilation rate, the CO_2 concentration remains low due to horizontal advection of poor CO_2 air mass from the southeast.

[29] The model analysis allows us to conclude on the importance of the combination of surface processes and mesoscale circulations in the ABL in order to establish precise budget of CO_2 at the regional scale.

[30] **Acknowledgments.** The authors would like to thank the referees for their constructive remarks and advices. The authors would also like to thank all the colleagues and CarboEurope partners who made CERES a very successful experiment and particularly the Météo France TRAMM and 4M teams as well as the SAFIRE staff for providing radiosounding, surface fluxes, and the Piper-Aztec data. Support for flying the Piper Aztec was mainly provided from CNES, CNRS-INSU, Météo-France and by MPI for the Dimona.

References

André, J.-C., J.-P. Goutorbe, and A. Perrier (1986), HAPEX-MOBILHY: A hydrological atmospheric experiment for the study of water budget and evaporation flux at the climatic scale, *Bull. Am. Meteorol. Soc.*, *67*, 138–144.

Bousquet, P., P. Ciais, P. Monfray, Y. Balkanski, M. Ramonet, and P. Tans (1996), Influence of two atmospheric transport models on inferring sources and sinks of atmospheric CO_2, *Tellus*, *48B*, 568–582.

Calvet, J., J. Noilhan, J.-L. Roujean, P. Bessemoulin, M. Cabelguenne, A. Olioso, and J.-P. Wigneron (1998), An interactive vegetation SVAT model tested against data from six contrasting sites, *Agric. For. Meteorol.*, *92*, 73–95.

Champeaux, J., H. Fortin, and K.-S. Han (2005), Spatio-temporal characterization of biomes over south-west of France using SPOT/VEGETATION and Corine Land Cover datasets, *IGARSS'05 Proc.*

Chevillard, A., U. Karstens, P. Ciais, S. Lafont, and M. Heimann (2002), Simulation of atmospheric CO_2 over Europe and Western Siberia using the regional scale model REMO, *Tellus*, *54B*, 872–894.

Denning, S., D. Randall, J. Collatz, and P. Sellers (1996), Simulations of terrestrial carbon metabolism and atmospheric CO_2 in a general circulation model. part 2: Simulated CO_2 concentrations, *Tellus*, *48*(4), 543–567.

Denning, S., M. Nicholls, L. Prihodko, I. Baker, P.-L. Vidale, K. Davis, and P. Bakwin (2003), Simulated variations in atmospheric CO_2 over a Wisconsin forest using a coupled ecosystem-atmosphere model, *Global Change Biol.*, *9*, 1241–1250.

Dolman, A., et al. (2006), CERES, the Carboeurope Regional Experiment Strategy in les Landes, South West France, May–June 2005, BAMS, in press.

Geels, C., S. Doney, R. Dargaville, J. Brandt, and J. Christensen (2004), Investigating the sources of synoptic variability in atmospheric CO_2 measurements over the northern hemisphere continents: a regional model study, *Tellus, Ser. B*, *56*(1), 35–50.

Gerbig, C., J. Lin, S. Wofsy, B. Daube, A. Andrews, B. Stephens, P. Bakwin, and A. Grainger (2003), Toward contraining regional scale fluxes of CO_2 with atmospheric observations over a continent: 1. Observed spatial variability from airborne platforms, *J. Geophys. Res.*, *108*(D24), 4756, doi:10.1029/2002JD003018.

Gurney, K., et al. (2002), Towards robust regional estimates of CO_2 sources and sinks using atmospheric transport models, *Nature*, *415*, 626–630.

Habets, F., J. Noilhan, C. Golaz, J. Goutorbe, P. Lacarrère, E. Leblois, E. Ledoux, E. Martin, C. Ottlé, and D. Vidal (1999), The ISBA surface scheme in a macroscale hydrological model applied in the Hapex-Mobilhy area, *J. Hydrol.*, *217*, 75–96.

Lafore, J., et al. (1998), The Meso-NH atmospheric simulation system. Part I: Adiabatic formulation and control simulations, *Ann. Geophys.*, *16*, 90–109.

Masson, V., J. Champeaux, F. Chauvin, C. Mériguet, and R. Lacaze (2003), A global database of land surface parameters at 1 km resolution for use in meteorological and climate models, *J. Clim.*, *16*, 1261–1282.

Nicholls, M., A. S. Denning, L. Prihodko, P.-L. Vidale, I. Baker, K. Davis, and P. Bakwin (2004), A multiple-scale simulation of variations in atmospheric carbon dioxide using a coupled biosphere-atmospheric model, *J. Geophys. Res.*, *109*(D18), D18117, doi:10.1029/2003JD004482.

Noilhan, J., and S. Planton (1989), A simple parametrization of land surface processes for meteorological models, *Mon. Weather Rev.*, *117*, 536–549.

Noilhan, J., P. Lacarrère, and P. Bougeault (1991), An experiment with and advanced surface parametrization in a mesobeta-scale model, *Mon. Weather Rev.*, *119*(10).

Rayner, P., I. Enting, R. Francey, and R. Langenfelds (1999), Reconstructing the recent carbon cycle from atmospheric CO_2, $\delta^{13}C$ and O_2/N_2 observations, *Tellus, Ser. B*, *51*(2), 213–232.

Schmitgen, S., H. Gei, P. Ciais, B. Neininger, Y. Brunet, M. Reichstein, D. Kley, and A. Volz-Thomas (2004), Carbon dioxide uptake of a forested region in southwest France derived from airborne CO_2 and CO measurements in a quasi-lagrangian experiment, *J. Geophys. Res.*, *109*(D14), D14302, doi:10.1029/2003JD004335.

Takahashi, T., R. Feely, R. R.W. ans Wanninkhof, D. Chipman, S. Sutherland, and T. Takahashi (1997), Global air-sea flux of CO_2: An estimate based on measurements of sea-air pCO_2 difference, *Proc. Natl. Acad. Sci. USA*, *94*, 8292–8299.

A. Butet, Service des Avions Français Instrumentés pour la Recherche en Environnement, Météo France, Toulouse, France.

F. Boumard, P. Ciais, J. D. Paris, and M. Ramonet, Le Laboratoire des Sciences du Climat et l'Environnement, Gif sur Yvette, France.

J. C. Calvet, S. Donier, C. Lac, P. Lacarrère, J. Noilhan, and C. Sarrat, CNRM/GAME, Météo France, 42, avenue Gaspard Coriolis, 31057, Toulouse, France. (claire.sarrat@cnrm.meteo.fr)

A. J. Dolman, Vrije University, Amsterdam, Netherlands.

C. Gerbig, Max Planck Institute for Biochemistry, Jena, Germany.

B. Neininger, MetAir, Menzingen, Switzerland.